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Recent developments in multiphoton strong-field physics

Man Mohan
Department of Physics and Astrophysics, University of Delhi, Delhi-110 007, India

Abstract : The interaction of intense laser with an atom involves the absorption or emission of many photon resulting excitation or ionization. Perturbation theories are quite successful in explaining various multiphoton experiments. When laser intensities, $I \gtrsim 10^{13}$ W/cm^2 and infrared frequencies are employed, then free electrons are dressed by interaction. Energies which exceed the photon energies. New non-perturbative multiphoton phenomenon then occur such as above threshold ionization (ATI) and higher harmonic generation (HHG). For higher intensities $I \gtrsim 10^{16}$ W/cm^2 the electron field of laser radiation becomes greater than the coloumb-binding potential of the outer shell electron of the atom, resulting over-the-barrier ionization. With recent progress in ultrafast-optics, the intensities of the laser has been continuously increasing and now the laser intensity up to $I \cong 10^{20}$ W/cm^2 is available. For such high intensities, stabilization of an atom against ionization is predicted recently and involves the formation of extended wave packet of the bound state which has little overlap with the nucleus and hence a low ionization rate.

Keywords : Multiphoton, intense Laser, excitation, ionisation, atom.

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1. Introduction

Recently there has been a rapid development in very high power lasers which has made available the entire range of intensities from less than 10^{-3} W/cm^2 to more than 10^{19} W/cm^2, an extraordinary range of some 22 orders of magnitude (or more) of controlled electromagnetic radiation, under laboratory conditions using chirped pulse amplification method [1]. It provides a unique opportunity for studying strongly perturbed dynamics of the simplest quantum systems.

At low intensity, the understanding of dynamical behaviour of interaction of radiation with matter has been developed in the context of the perturbation theory. Thus for example not only linear but also the non-linear multiphoton process could be investigated and understood within the framework of the ordinary perturbation theory, albeit with due generalization of the usual first order theory of the n-th order non-vanishing order (for an photon process). Multiphoton processes (*e.g.* multiphoton excitation and ionization) results from simultaneous absorption of several photons. Multiphoton transition were first predicted by Goppert-Mayer in 1931 [2] and observed at radio frequencies by Hughes and Gralner in 1950 [3]. The study of multiphoton absorption at optical frequencies only became possible when intense laser sources were developed during early 1960. Following this, the two photon excitation in CaF2 was first observed by Kaiser and Garrett in 1961 [4] and later two photon excitation of cesium was demonstrated by Abella [5].

2. Multiphoton excitation

These days two-photon spectroscopy, especially through intennediate resonances, has become a standard tool in atomic and molecular photophysics [6]. A resonance enhancement of many orders of magnitude has been observed in all alkali atoms when the frequency of one of the photons corresponds to the energy of an intermediate state.

In spectroscopy, the study of H-atom plays a great role, as exact calculations can be perfonned and bench mark comparison can be made between theory and experiment. So far, only two photon transition between the ground state to 2s state has been studied both experimentally [6.7] and theoretically [8]. Further with the development of Synchroton radiation and laser sources around the world, it has now

become possible, in principle, to measure two-photon absorption coefficients of hydrogen transitions between different excited states. For this reason, the evaluation of two photon transition probabilities accurately has become timely and very important.

Generally perturbation methods are employed in order to describe multiphoton processes. When N-th order time dependent perturbation theory is applied, a major difficulty found in such calculations is the infinite summation over the complete set of unperturbed atomic states, discrete plus continuum, related to the transition matrix of the perturbation theory. Earlier several methods were used in order to evaluate these sums. Bebb and Gold [9] defined an average frequency $\bar{\omega}$ thereby replacing the infinite summation by an average term and using the closure property of the wavefunctions to remove the intermediate state transitions from the problem. Gontier and Trahin [10] related the infinite summations to the numerical solutions of a set of first order differential equations. Laplance *et al* [11] used the Green's function method to perform the summations.

Recently, we have calculated absorption coefficient for the two photon process, which involves evaluation of summation over intermediate states [12]. We have replaced the intermediate states by a finite set of pseudostates which are expanded in terms of a basis which is descrete and complete. Using these pseudostates we have found excellent convergence even for a small basis size. The main advantage of the method is that it replaces the infinite summation over both discrete and continuum states by a finite sum over the pseudostates. The energy spectrum of the pseudostates contains both negative and positive values representing both the bound states and the continuum states adequately.

2.1. The pseudostate method :

In the pseudostate method we diagonalize the target Hamiltonian in terms of a basis which are discrete and complete and expand the target wave functions in terms of the these basis functions. The atomic unperturbed Hamiltonian for the hydrogen atom is (in Rydberg units).

The basis functions taken are of the simple fonn [13]

$$\varphi_j = e^{-ar} r^{l+j} Y_{lm}(\vartheta, \varphi), \quad j = 1, 2, \ldots N \tag{1}$$

where a = basis parameter

l = orbital angular momentum

N = size of basis

The target wave functions are expanded in terms of the linear combinations of the basis functions as

$$|n\rangle = \Psi_n = \sum_{j=1}^{N} C(j,n)\varphi_j / r \tag{2}$$

and we have

$$\langle n|H_0|n'\rangle E_n \delta_{nn'}, \quad \langle n|n'\rangle = \delta_{nn'} \tag{3}$$

where H_0 is unperturbed Hamiltonian of the atom.

On diagonalising the target Hamiltonian in basis of size N one obtains N energy eigen values spanning both positive and negative energies. The lowest lying negative energy eigenfunctions are excellent approximations to the exact bound state wave functions while rest of them represent in some way all other bound states. The pseudostate energies which have positive values represent adequately the continuum states of the atom.

Once we know the eigenvalues and eigenvectors one can solve the integrals involved for evaluation of the transition amplitudes. Since the number of basis functions required for proper convergence is small the Pseudo State Method is a simple and powerful technique to be used in calculations where one has to take the infinite number of target wavefunctions into account.

The two photon transition probability amplitude D from an initial state $|i\rangle$ to a final state $|f\rangle$ in the length gange is given by

$$D = 3/2 \sum_{\bullet} (1 + P_{12}) \left[e_1 \cdot \langle f|r|n\rangle \langle n|r|i\rangle \cdot e_2 / (\nu(n) - \nu(i) - \nu_2) \right] \tag{4}$$

where e_1 and e_2 are the polarisations of the incident photons and the operator P_{12} interchanges the frequency and polarisation of the two photons and $\nu(n)$ are the demensionless frequencies of the intermediate states given by

$$\nu(i) = E_i / 2\pi\hbar R \tag{5}$$

here R is Rydberg frequency and is equal to 3.29×10^{15} per second.

In order to see the accuracy involved in our calculations we have compared our results for the $1s - 3s$ transition in hydrogen with those obtained by Bassani *et al* [8]. The results are obtained by two entirely different methods. Bassani *et al* have used the standard radial eigen functions of hydrogen for both bound states and the continuum while we have approximated all the eigen functions of the unperturbed hydrogen in terms of the linear combinations of the functions of a discrete basis of small size. As can be seen from the Table 1 our results in Kundilia, Prasad and Mohan [12] are in excellent agreement with those of Bassani *et al*. As very few basis functions are needed for the proper convergence of the results one can easily calculate the transition amplitudes without much computational effort.

Table 1. Three-photon ionization cross section (cm^6/W^2) of $H(1s)$ as compared with the results of others. Figures in the bracketts indicate power of ten.

λ(Å)	LDFJR[a]	GS[b]	Present	LDFJR[a]	GS[b]	Present
	Linear polarisation			Circular polarisation		
1900	1.186(–46)	1.172(–46)	1.150(–46)	2.479(–46)	2.549(–46)	2.680(–46)
2000	5.581(–48)	5.429(–48)	5.426(–48)	1.365(–47)	1.326(–47)	1.327(–47)
2100	2.542(–47)	2.541(–47)	2.540(–47)	5.771(–47)	5.776(–47)	5.771(–47)
2200	1.593(–47)	1.589(–47)	1.583(–47)	3.957(–47)	3.945(–47)	3.931(–47)
2300	2.650(–47)	2.641(–47)	2.614(–47)	3.853(–47)	3.840(–47)	3.818(–47)
2400	7.125(–46)	7.057(–46)	6.854(–46)	3.935(–47)	3.917(–47)	3.882(–47)
2500	2.980(–46)	2.948(–46)	2.942(–46)	3.998(–47)	3.972(–47)	3.935(–47)
2600	1.008(–46)	1.002(–46)	0.993(–46)	3.947(–47)	3.906(–47)	3.881(–47)

[a]Laplanche *et al* (1976); [b]Gao and Starace (1988).

On calculating the transition amplitude one can easily calculate the transition rate (TR) and the absorption coefficient. The absorption coefficient is given by

$$\alpha(\omega_1) = WhN_0 \Big/ \left\{0.5\varepsilon_1\varepsilon_0 |E_1| 2c/\sqrt{\varepsilon_1}\right\}$$

In the above expression N_0 is the density of atoms in the sample; ε_1 is the dielectric function of the medium for radiation of frequency ω_1; W is the two photon transition probability per unit per atom, given by

$$W = |E_1 E_2|^2 e^4 a_0^4 |D|^2 \delta(\Delta\nu) \Big/ \left\{36R^3(2\pi h)\right\}$$

where E_n and E_2 are the electric fields of the incident electromagnetic wave; a_0 is the Bohr radius; $R = 3.29 \times 10^{15}$ s^{-1} is the Rudberg frequency and $\Delta\nu = \nu(f) - \nu(i) - \nu 1 - \nu 2$ gives the energy conservation. $|D|^2$ is defined as the dimensionless two-photon absorption coefficients (DTAC).

3. Multiphoton ionization

Multiphoton ionization (MPI) in which electron is ejected after absorption of many photons was first observed in the experiments of Hall, Robinson and Branscomb, in which ruby laser was used to induce two photon detachment from negtive halgon ions [14]. Later MPI from rare gas atom was observed by Voronov and Delone [15] and Agostini *et al* [16].

The multiphoton ionization of an atom reflects both the characteristics of the laser pulse and the properties of atom perturbed by the intense laser field. Multiphoton ionization thus constitutes a very favourable method for studying the response of an atom in the presence of an intense laser field. Recently we investigated [17] the multiphoton ionization for atomic hydrogen in the ground and excited sates using powerful pseudo-state summation technique.

In Table 1 we have compared our results with the results obtained tly Laplanche *et al* [15] by Gao and Statace [16] using variational technique. As can be seen our results obtained by pseudo-state method is quite agreement with others results. In Figure 1 we have shown the variation of three-photon ionization rate from the ground as a function of wave length of incident photons. Results shows the enhancement of cross sections near the intermediate resonances, where the detuning of laser frequency with respect to real bound state becomes quite small.

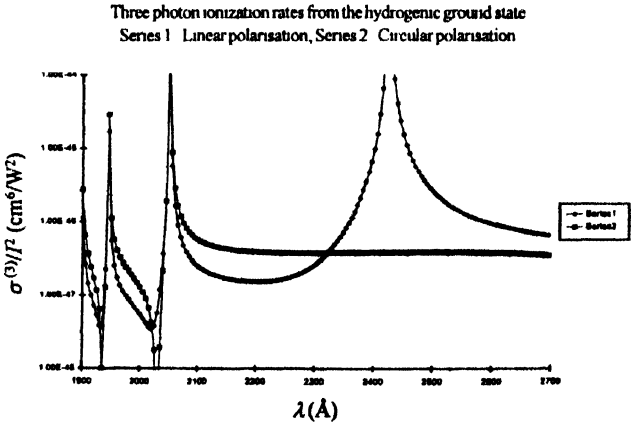

Figure 1. Variation of three-photon ionization rates from the hydrogenic ground state as a function of wavelength of incident photons. Here we have considered both linearly and circularly polarised light.

As stated earlier, due to the availability of laser sources of very high intensities these days, there is a substantial shift in research activities in multiphoton processes. Other exciting main emerging areas are above threshold ionization, the production high order harmonic generation and stabilization in high intense laser field *etc*.

4. Above Threshold Ionization (A.T.I.)

For a long time, there was a commonly held picture that in N photon ionization of an atom, that the electron would move away from the ion with a kinetic energy $Nhw-Ei$, where Ei is

the atomic ionization energy, hw the photon energy and N is the minimum number of photons required to reach the ionization threshold. This is a simple multiphoton extrapolation of the Einstien picture of the photoelectric effect. As a result, the electron energy distribution is expected to display a single electron peak centred at an energy Nhw–Ei. This picture prevails at weak laser intensity.

However, an experiment on 6-photon ionization of xenon atoms performed at 10^{12} W cm^{-2} was shown that, addition to the expected electron peak, a second one separation from the first by a photon energy was observed [18]. The presence of this second peak means that the electron has absorbed more energy than consistant with the minimum numbers of N of photons. This effect which was named as above threshold ionization (A.T.I.), became new and important sub-field of multiphoton ionization of atoms. A.T.I. has been observed in different atoms and molecules, and laser in the infrared, visible and ultraviolet. The electron energy spectrum consists of a series of peaks evenly spaced by the photon energy. It corresponds to the absorption of $N + S$ photons with large s numbers. The ionization continuum, which was devoid of any structure in a weak laser field thus acquires a structure of states induced by the intense laser field, this stucture corresponding to electron peak appers at harmonics of the laser frequency [18].

5. Higher-order harmonic generation in atoms

A laser radiation which interacts with an atom induces a dipole moment in the atom which in turn acts as a source of radiation. At higher intensities $I > 10^{13}$ Watts/sq cm the atomic response becomes nonlinear and as a result new frequencies appear at multiples of the driving frequencies. It was pointed out by Shore and Knight that high-order odd-harmonics generation could take place by the emission of high energy continuum state and a low lying bound state [19]. Subsequently very high order odd-harmonics generation had been reported in rare gases using a 1064 nm Yag laser at an intensity of 10^{13}–10^{14} W/sq cm (Perry *et al* [20]). They produced spectra that exhibited what have now been recognized as a typical characteristics for harmonic spectra. Main observable characteristics in this area rapid decrease in the harmonic intensity from third to seventh order; a plateau region that extends out to 27-th order, with harmonics having similar intensities; and a rapid drop in harmonic strength beyond the 27-th order. These features have also been observed in the spectra of many other atoms [21]. Most of the calculations, even with crude models, are able to reproduce only qualitatively the experimental results [21] with the decrease in efficiency for the first harmonics, the plateau and the cutoff. This may be due to very general property of a strongly driven non-linear system. One of the interesting questions regarding the harmonic generation spectra concerns the nature and exact location of the cutoff. The cuttoff location sets the ultimate limit for the highest frequency that can be efficiently generated. Obviously, this question is of great importance from the point of view of possible applications for the production of VUV intense radiation sources. Numerical calculations of Krause *et al* [22] are shown that the maximum energy at the end of the plateau is well approximated by the simple and universal formula $I_p + 3E_p$, where I_p is atomic ionization potential and E_p is the pondermotive energy in the laser field of strength E and frequency w. The cuttoff in the harmonic spectrum occurs for harmonics of order higher than

$$N_{max} \cong (I_p + 3E_p)/w.$$

The overall maximal photon energy (in units of W) that can be achieved is then approximately given by the value of this expression at the saturation intensity Isat at which the atom ionizes. However, the above equation can further be modified when collective effects (phase matching) become relevant. The classical simulation [23] show that the maximum kinetic energy acquired by free electrons from the field when they return to the nucleus 3.2 E_p which is close to the prediction of [24]. The classical interpretation shows that in order to control harmonic generation processes, one should try to control the motion of free electrons in the laser field. Shaping approximately electron trajectories might allow for various fascinating applications, including, for example, the generation of sub-pico second high generation pulses.

6. Suppression of ionization

Recently, attention has been focussed on the issue of stabilization and suppression of ionization in an intense laser field [25–28]. Using this process one will be able to expose neutral atoms to high intensity laser fields. Various mechanisms have been suggested that could bring about ionization suppression. We can broadly divide these into two types. The first type we term as quantum interference suppression, because the mechanisms of this type are generally interpreted in terms of destructive interference between the ionization from closely spaced bound states [26,27]. The second general type is a subject of tropical interest at present and can be labelled as super intense ionization suppression. Such suppression typically occurs in fields of $I > 10^{16}$ W/sq cm or greater and has to do with the production of electronic wave packets which oscillate in the laser field with large amplitudes far from the nucleus with diminished ability to absorb photons [25,26]. In conclusion, super intense ionization suppression is a phenomenon of great interest as it will be testing ground for various non-perturbative theories which are developing. Further this process has stimulated more interest as it is completely opposite to our basic concepts that strong laser fields produce rapid evolution.

Conclusion

Here in this work we have covered the whole range of developments in multi-photon physics which advancement of laser technology in recent years.

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